

Title: Investigation of a Novel Catalyst for NO Decomposition

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Industrial Collab.: Airflow Catalyst Systems

Grant Number: DE-FG26-03NT41911

Performance Period: September 15, 2005– April 30, 2006

ABSTRACT

Objective:

The main objective of the proposed research is the evaluation of the Pt/SnO₂ catalysts for the decomposition of NO in simulated power plant stack gases with particular attention to the resistance to deactivation by O₂, CO₂, and elevated temperatures.

Accomplishments to Date

In the previous reporting period temperature programmed reaction runs on the 15% Pt and 10% Pt catalysts were performed using NO+He and NO+O₂+He reactant gas mixtures.

The catalyst was active for NO decomposition only above 600 K and at these temperatures no appreciable N₂O was observed. For feeds containing additional oxygen, the NO decomposition activity was less when fresh and the catalyst lost most of its activity in the subsequent runs. This may be due to the oxidation of Pt, coverage of active Pt sites with oxygen, and the loss of OH groups from the SnO₂ surface. Since these OH groups are thought to be very active in the O₂-involving processes on the catalyst surface, and since water vapor is a component of the power plant stack gas, we plan to repeat the TPRx runs with feeds containing water vapor.

Temperature programmed reaction of NO + O₂ + H₂O on 15 % Pt/SnO₂ catalyst was also done. The reaction of NO and O₂ in the presence of water vapor appears to show two different reaction schemes, one below and the other above 750 K. Activation energy below 750 K was estimated to be 204 kJ/mol and that at higher temperatures was estimated as 43kJ/mol.

In addition, reactions of dry and wet mixtures of NO and O₂ were carried out for over 3 hours. No apparent deactivation was observed.

In this reporting period the focus was on establishing the effects of pretreatment, presence of oxygen and water vapor in the feed, and space velocity. Runs were made at 1000 K and 900 K to establish the variation of NO conversion with temperature. The Temperature Programmed Reaction (TPRx) runs were performed using the Micromeritics Pulse Chemisorb 2705 with TPD/TPR Option. A mass spectrometer-gas chromatograph system (SATURN 2000MS/3800GC) from Varian was used for the identification of the reaction products. The calibration of the GC-MS was completed and the results for this period were reported as concentration of species in ppm.

Two different pre-treatment methods were used. One at 373 K under flowing He for 2 hours mainly to drive off the air and physically adsorbed gases, the other at 900 K under flowing He for 2 hours to drive off most of the OH groups attached to the tin oxide that contribute to the non-specific adsorption of emission effluent species that might mitigate the catalytic activity. The results show that this effect is in fact present and the NO conversion is higher on the catalyst that has been treated at high temperature (13 % compared to 9 % obtained in the presence of O₂, on the catalyst treated at low temperature).

The results show that the presence of equimolar amount of O₂ drastically reduces the conversion from 30 % to 13% for reaction at 1000 K. The activity of the catalyst increased with reaction temperature, mainly due to the fact that the thermal desorption of oxygen is facilitated at high temperatures, regenerating the active sites of the catalyst. Water in the feed had no effect on NO conversion under the reaction conditions in this study. This was an unexpected result and may be due to the fact that water and NO adsorb on different sites of the catalyst.

Comparison of the results of 40 ccpm and 60 ccpm feed rates indicate that increase of gas velocity and the space velocity does not significantly affect the NO conversion.

The BET surface areas of the 15 % Pt and 10 % Pt catalysts were also measured.

Future Work

In the remainder of the project the effect of CO₂ will be investigated and long duration runs will be made to establish the deactivation characteristics of the catalyst. A final comprehensive report will be produced.

Additional Activities

- A paper, entitled “Nitric Oxide Decomposition on Pt/SnO₂ Catalysts” was submitted for presentation at the 2006 AIChE Annual Meeting.
- Jillyan Harlan and Taikelia Battle, both undergraduates, were supported under this grant